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FALL-OUT PHENOMENOLOGY

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San Francisco 24, California
August 1951

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length of time being roughly proportional to the activity on each slide. After development, the slides were examined and a number of exposed spots on the film were found. By using above-stage lighting, it was possible to identify a radioactive particle in or near the center of most of the spots.

The advantage of this method is that it enables the study of the active particles in an undisturbed state as they fell onto the fall-out plates. There is no danger of disrupting their structure or breaking down agglomerates as there is in the sedimentation procedure. The disadvantage of the method is that it is not applicable to particles less than 0.5 μ due to the limitation of the optical microscope and to the relatively low level of activity of small particles necessitating a very long exposure during which the film is blackened excessively by the larger, more highly radioactive particles.

A microscopic study of these slides showed that the radioautographs were almost exclusively associated with black spheres. These black spheres ranged in size from about 450 μ in diameter to submicroscopic dimensions.

The spheres showed three modes of occurrence: (1) as individual particles, (2) as parti-

TABLE 3.2 RELATIVE RADIOACTIVITIES OF FALL-OUT SIZE FRACTIONS

SHOT	ISLAND	SIZE OF FRACTION (μ)	WEIGHT OF FRACTION (mg)	TOTAL COUNTS[a] PER MIN PER PLATE × 10^{-5}	PER CENT OF TOTAL ACTIVITY
		<2	0.4	0.35[b]	0.5
Dog	Aniyaanii	2–20	2.3	2.4	4
		>20	142.2	57.0	95.5
		<2	4.8	1.40[c]	3
Dog	Parry	2–20	6.3	6.0	14
		>20	113.3	36.5	83
		<2	1.1	4.4[b]	12
Dog	Eniwetok	2–20	4.8	4.2	12
		>20	1,809.8	27.3	76
		<2	1.0	0.87[b]	2
Dog	Igurin	2–20	3.9	6.1	13
		>20	1,952.9	40.2	85
		<2	0.6	15.9[b]	6.5
Dog	Giriinien	2–20	2.0	15.9	6.5
		>20	1,133.9	214.0	87
		<2	0.7	59.1[b]	8
Dog	Rigili	2–20	3.8	96.9	14
		>20	39.9	549.5	78
		<2	1.6	152[b]	1
Easy	Bogallua	2–20	17.6	1,510	7
		>20	160.4	19,300	92
		<2	2.7	17.8[b]	1
Easy	Kirinian	2–20	2.5	76.2	6
		>20	44.2	1,380	93
		<2	0.9	6.8[d]	0.5
Easy	Piiraai	2–20	5.6	41.5	3.5
		>20	49.5	1,156	96

[a] The total counts per minute have been corrected for decay back to shot time plus 10 hr by means of the decay curves for Dog and Easy Shots obtained from Project 6.1.

[b] Geometry of counter is 3 per cent as determined by use of a RaDE standard.

[c] Geometry of counter is 2 per cent (RaDE standard).

[d] Geometry of counter is 8 per cent (RaDE standard).

cles adhering to coral grains, and (3) in loosely associated clusters of spheres and granules of a transparent material.

Approximately one-third of the radioactive particles found on the slides were individual spheres. These spheres were almost perfectly round and superficially homogeneous. They ranged from about 1 to 20 μ in diameter. They were found in all samples from both Dog and Easy Shots. Figures 3.2 and 3.3 show the size distribution of a number of spheres measured in samples collected on Rigili and Bogallua Islands. Many of the Easy Shot samples showed that a high percentage of the activity was due to individual spheres or subspherical particles 200 to 450 μ in diameter. These spheres exhibited a granular texture under the microscope. Their surfaces were irregular and had a black, glossy, beaded appearance. Very small grains of what appeared to be calcium carbonate were adhering to the surfaces. (The grains had very high birefringence, characteristic of calcium carbonate, under the petrographic microscope.) Some of the spheres had cracks and cavities, the appearance of which suggested that the granular structure persisted throughout the interior of the sphere. A number of these large spheres were removed from a fall-out plate with tweezers, and their diameters and radioactivity measured. There was no relationship between the sizes of the particles and their activities. One sphere was found that counted approximately 3×10^6 disintegrations/sec (corrected to E+10 hr).

Most of the black spheres, especially in the Dog Shot samples, were found adhering to coral grains. The sizes of the black spheres so found varied from about 10 μ in diameter to submicroscopic. The number of adhering spheres per grain varied from 2 or 3 to perhaps 50. The sizes of the coral grains themselves varied from about 10 to 500 μ in diameter. A large number of these radioactive grains from the Rigili and Bogallua samples were measured, and their size distribution is shown in Figs. 3.4 and 3.5. Where these grains were thin enough to be viewed with transmitted light in the petrographic microscope they exhibited the characteristic very high birefringence of calcium carbonate. There were a large number of salt crystals visible in the slides, none of which were observed to carry radioactive particles. In one instance, a vegetable fiber was found which had a large number of radioactive spheres on the order of 1 or 2 μ in diameter adhering to it. Only a very small fraction of the total number of particles found on the slides carried radioactive particles.

In quite a few cases the active spheres were found in very loose, irregular aggregates or clusters intimately mixed with an amber-colored, isotropic, glassy material. Parts of the aggregate were separated from each other. Apparently the particles fell as loosely bound floccules which shattered when they hit the fall-out plate.

All the data on the measurements of radioactive particles by the radioautographic technique have been compiled in Appendix A, Table A.1.

3.2.3 Chemical Analysis of Fall-out Samples

Six samples of the Easy Shot fall-out from Bogallua were analyzed for the radioactive elements present. The six samples were:

(1) An unfractionated sample removed directly from the fall-out plate with xylene and concentrated by centrifugation.

(2) The xylene supernate from this centrifugation.

(3) A sample of the large radioactive spheres picked with tweezers from a fall-out plate.

(4) The small fraction from a sample which had been size-fractionated by the sedimentation method.

(5) The medium-size fraction.

(6) The large-size fraction.

The results of the analyses are shown in Table 3.3.

Several of the large spheres, 200 to 450 μ in diameter, were analyzed spectrographically to determine the elements which make up the bulk of the particles (see Table 3.4).

In addition, an attempt was made to identify the composition of the large spheres by X-ray diffraction. A powder photograph showed a series of lines which indicated the composition of the particles to be about one-third calcium carbonate (calcite), one-third calcium oxide, and one-third unidentified compounds.

3.2.4 Radioautographs of Fall-out Plates

Radioautographs of many of the fall-out plates were made shortly after their arrival at USNRDL. A typical one is reproduced in Fig. 3.6. This radioautograph was made from a fall-out plate from Parry Island (Dog Shot) with Eastman Type K X-ray film enclosed in two lighttight envelopes. The packaged film was pressed against the fall-out plate with sponge rubber, plywood, and a lead brick, and was exposed for 5 hr on D+7 days.

By means of another series of exposures, the radioactivities associated with two of the spots were determined with a densitometer designed for small areas. The activities of spots A and B (Fig. 3.6) were found to be 14 and 24 rep/hr, respectively (at the surface of the fall-out plate), on D+9 days.

TABLE 3.3 RADIOCHEMICAL ANALYSES OF EASY SHOT FALL-OUT

ELEMENT	UNFRACTIONATED SAMPLE		XYLENE WASH		LARGE SPHERES 200–450 μ IN DIAM	
	Found (%)	Theoretical (%)[a]	Found (%)	Theoretical (%)	Found (%)	Theoretical (%)
Total rare earths—La, Pr, Nd, Ce, Y	63	60	79	58	60	62.5
Ba	5.6	11.8	6.3	8.9	6 (Ba + Sr)	21.5 (Ba + Sr)
Sr	0.7	10	....	....		
Zr	16.3	9	3.2	11.6	16.6	8
Ru	5.5	4	3.8	4.7	9.0	4
TOTAL	91	95	92	83	92	96
Time of Analysis	E+39 days		E+53 days		E+33 days	
	Small Fraction, Less than 2μ		Medium Fraction, 2 to 20μ		Large Fraction, Greater than 20μ	
Total rare earths	82	59	88	60	73	60
Ba	5	12	11.2 (Ba + Sr)	22 (Ba + Sr)	3.6	12.6
Sr	....	....			0.1	8.7
Zr	12	9	....	....	14.1	7.3
Ru	....	....	....	....	5.0	5.8
TOTAL	99	80	99	82	96	94
Time of Analysis	E+39 days		E+35 days		E+27 days	

[a] The calculated theoretical percentages are determined from considerations of the fission-yield report, USNRDL Report ADC-65, H. F. Hunter and N. E. Ballou, *Simultaneous Slow Neutron Fission of U^{235} Atoms*, and the recent Greenhouse data on fission yields.

TABLE 3.4 SPECTROCHEMICAL ANALYSIS OF LARGE FALL-OUT PARTICLES

ELEMENT	STRENGTH OF LINES	
Al	W	
Ca	VS	
Cu	W	
Fe	M	T=0.001–0.01%
Mg	T	W=0.01–0.1%
Mn	T	M=0.1–1%
Si	S	S=1–10%
Sn	T–W	VS=>10%
Ti	S	
Zn	T–W	

Chapter 4

Discussion

4.1 RELIABILITY OF EXPERIMENTAL PROCEDURE

The results of the sedimentation procedure indicate a moderate amount of radioactivity on particles in the small- and medium-size fractions from the Dog Shot samples. These results differ consistently from the results of the Easy Shot fall-out analysis which indicate that practically all of the radioactivity is associated with the large fraction.

As it might be expected from a consideration of the settling velocities of particles in air that the fall-out should consist of large particles only, the apparent high percentage of active small particles in the Dog Shot fall-out is surprising. The question arises whether the small particles actually fell to the ground individually or whether they fell attached to larger grains and were dislodged during the sedimentation procedure.

The removal of the particles from the fall-out plates and the separation of the large fractions from the small and medium fractions were done with the particles suspended in xylene. Xylene, being a hydrocarbon whose molecule contains no polar groups, does not tend to disperse particles suspended in it. It does not seem likely that a significant dispersion of the particles occurred in these stages.

During the process of separation of the medium from the small fractions, the samples were suspended in water and subjected to ultrasoneration. At this point it is probable that an appreciable dispersion of the particles occurred which would result in an increase in the number of radioactive small particles at the expense of the medium-size particles.

From these considerations considerable doubt is cast upon the accuracy of the fractionation in the smaller size ranges. That particles in the medium-size range actually fall out individually is, however, borne out by the results of the microscopic examination of the radioautographs in which single particles down to 1 μ in diameter were found.

Very few radioactive particles smaller than 2 μ were found by this method. This indicates that most of the radioactivity of the small fraction was due to the small radioactive spheres removed from the large coral grains during the sedimentation procedure.

4.2 ORIGIN OF THE FALL-OUT PARTICLES

It seems probable that the black, radioactive spheres originated by condensation of material from the ball of fire within a short time after the shot. The only evidence available as to the chemical composition of the particles comes from the spectrographic analysis of the large (200 to 450 μ) spheres from the Bogallua fall-out after Easy Shot. The major constituents are calcium, silicon, iron, and titanium. The calcium must have originated from the vaporization of large amounts of coral (almost exclusively calcium carbonate) near the base of the shot-tower. The iron and silicon probably came from the vaporization of the steel tower and its concrete foundation. A source of large amounts of titanium is unknown. The results of the X-ray diffraction study of these spheres show that the calcium is in the form of the carbonate and the oxide. It seems likely that the calcium originally condensed as the oxide and by reaction with carbon dioxide in the air has been partially converted to the carbonate. No evidence of calcium hydroxide was found. The carbonate shows the crystal structure of calcite in contrast to the unaltered coral sand which has the aragonite structure.

Examination of the radiochemical analysis of the fall-out shows that fractionation of the active elements has occurred during the formation of the particles. The relative amounts of strontium and barium are consistently less, in comparison with the other radioactive elements reported, than would be expected from the normal fission-yield distribution. An examination of the fission-product chains[1] shows that the strontium isotope most likely to be found in the radiochemical analysis has Kr^{89} with a 2.6-min half life for an ancestor. Most of the Kr^{89} formed in fission would not have decayed at the time of formation of the spherical particles. Similarly, barium has as an ancestor Xe^{140} with a 16-sec half life. From the half lives of their gaseous ancestors, it would be expected that there would be considerably less strontium in the fall-out particles than barium. This is actually the case. The amount of strontium reported varies from about 1 to 7 per cent of what would be expected from the normal yield, while the amount of barium present is about 50 per cent.

As previously described, most of the fall-out particles collected consist of inert coral grains with several small, radioactive spheres adhering. It does not seem likely that the spheres became attached to the sand grains after the spheres reached the surface of the ground. In that case it would be expected that the spheres would be more or less evenly distributed over all the surface grains, and that there would be a high percentage of free spheres in the fall-out collectors. Such is not the case. The number of active grains relative to the number of inactive grains is very small, as is the number of free spheres relative to the number of active grains. It seems probable that the spheres became attached to the grains in the air after the spheres had formed and the ball of fire had cooled to a relatively low temperature. The coral grains show no indication of having been fused, and in one case the spheres are found adhering to a vegetable fiber. Probably a considerable amount of surface debris was carried aloft by the strong updrafts following the ascent of the ball of fire. Many of the small spheres formed in the radioactive cloud must have adhered to this debris which subsequently reached the ground as fall-out.

4.3 MECHANISM OF FALL-OUT

Stokes' law is of value in explaining the mechanism of, or predicting, fall-out. However, one of the basic assumptions of the law is streamline, nonturbulent settling, which condition certainly does not prevail in the vicinity of an atomic blast. Quite a number of individual radioactive particles 5 μ in diameter were found in the radioautographs of the fall-out. According to Stokes' law it should take a spherical 5-μ particle about 4 weeks to fall from 20,000 ft to sea level (assuming a particle density of 3.0). It is obvious that the downward velocity of the particles must be affected by some mechanism other than mere gravitational settling.

It is known that during the development of cumulus clouds, where strong updrafts rise rapidly to heights of many thousands of feet, there are associated weaker down currents of air somewhat after the manner of convection currents. These downdrafts occur around the central rising current and extend out to a distance of several times the diameter of the rising current. It seems possible that there could be some such development of down currents in the vicinity of the rising atomic cloud which could give to a swarm of particles a downward velocity *much greater than their normal settling velocities.* In such a case, the particles would be carried downward with a velocity independent of their size, and the resulting fall-out would show much less size segregation than if it were due to Stokes'-law settling alone.

[1] Plutonium Project, "Nuclei Formed in Fission," *J. Am. Chem. Soc.*, LXVIII (1946), 2411.

Appendix A

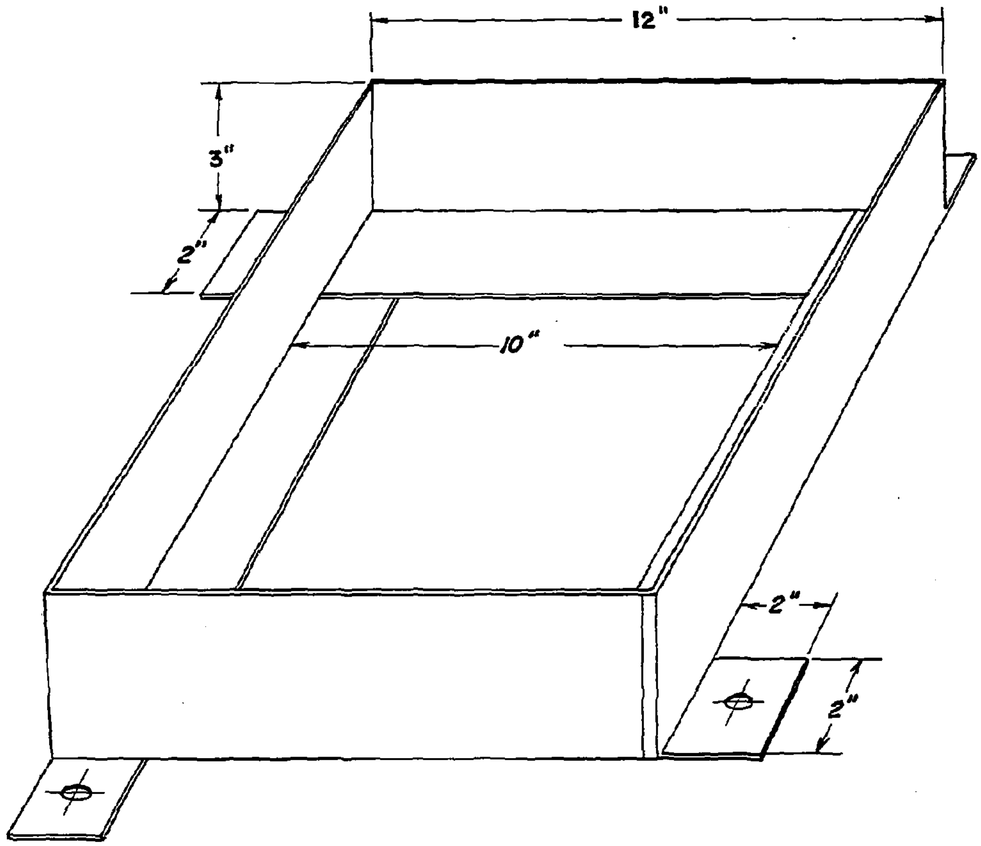
Size Distribution of Radioactive Fall-out Particles

Table A.1 is a compilation of measurements of all the radioactive particles found with the radioautographic technique described in Section 3.2.2. The figures in the table are the numbers of particles whose diameters fall within a given size range.

Figures 3.2, 3.3, 3.4, and 3.5 were drawn using data from this table.

TABLE A.1 SIZE DISTRIBUTION OF RADIOACTIVE FALL-OUT PARTICLES

	EASY SHOT				DOG SHOT									
	Bogallua		Kirinian		Aniyaanii		Parry		Eniwetok		Igurin		Rigili	
DIAMETER (μ)	Individual Spheres	Active Coral Grains	Individual Spheres	Active Coral Grains	Individual Spheres	Active Coral Grains	Individual Spheres	Active Coral Grains	Individual Spheres	Active Coral Grains	Individual Spheres	Active Coral Grains	Individual Spheres	Active Coral Grains
0.0 – 1.0	1	..	..	..	..	..	..	..	..	..	...	..	..	..
1.0 – 1.25	2	..	..	..	..	..	..	..	..	..	..	..	..	..
1.25– 1.6	3	..	1	..	..	..	..	..	..	..	..	..	5	..
1.6 – 2.0	12	..	1	..	1	..	..	..	..	..	1	..	8	..
2.0 – 2.5	11	..	2	..	1	..	..	..	..	..	1	..	10	..
2.5 – 3.2	18	2	..	1	1	..	1	..	..	..	3	..	21	..
3.2 – 4.0	40	4	5	..	2	..	1	..	3	..	2	1	22	1
4.0 – 5.0	40	11	1	2	4	3	1	..	2	..	4	..	27	3
5.0 – 6.3	59	23	7	2	2	..	1	1	3	..	1	..	23	5
6.3 – 8.0	53	36	9	5	1	..	1	..	2	1	1	1	11	14
8.0 – 10.0	28	43	1	3	3	4	..	1	..	2	1	2	8	9
10.0 – 12.5	18	52	1	6	..	4	..	1	1	..	2	4	2	10
12.5 – 16.0	19	60	..	5	1	1	..	1	..	..	1	1	..	23
16.0 – 20.0	6	60	..	10	..	1	..	1	1	2	..	1	..	15
20 – 25	1	65	..	7	..	1	..	..	..	2	..	..	1	19
25 – 32	1	59	..	3	1	..	..	3	..	2	..	..	..	21
32 – 40	3	37	..	5	..	1	..	..	..	2	..	1	1	16
40 – 50	..	33	..	5	..	..	..	..	..	3	..	1	..	11
50 – 63	1	30	..	5	..	..	..	2	..	4	..	..	..	4
63 – 80	..	18	..	4	..	1	..	2	..	2	..	..	1	10
80 – 100	..	15	..	5	..	..	..	3	..	3	..	..	..	4
100 – 200	..	29	..	19	..	4	..	9	..	7	..	1	..	13
200 – 500	..	12	..	5	..	4	..	2	..	6	..	4	..	4
500 –1000	..	8	..	..	..	..	..	..	..	..	..	..	..	..
TOTAL	316	597	28	92	17	24	5	26	12	36	17	17	140	182



MATERIAL - 1/32" SOFT ALUMINUM PLATE

FIG. 2.1 Fall-out Plate Holder

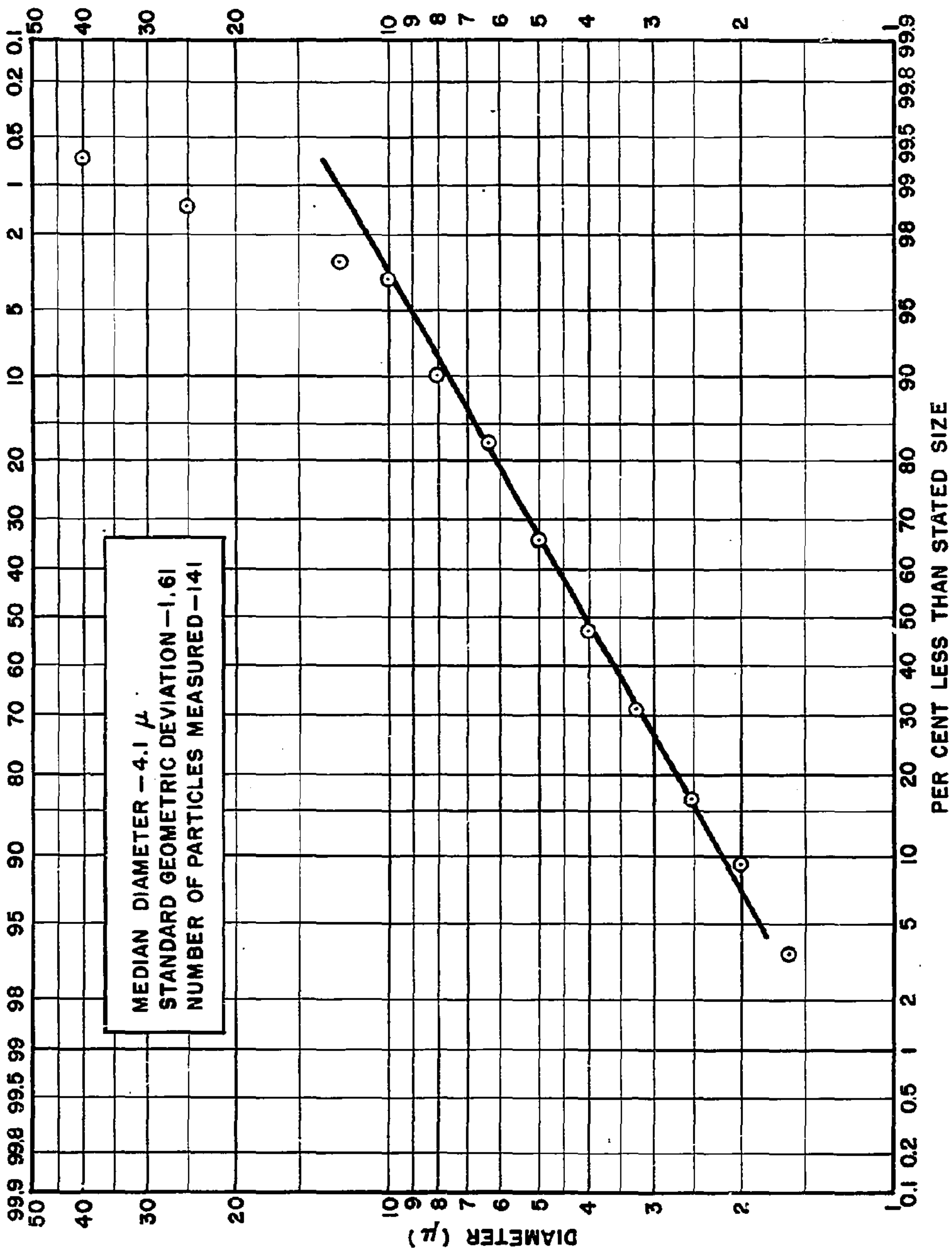


FIG. 3.2 Size Distribution of Individual Radioactive Spheres Found in Samples from Rigili Island (Dog Shot)

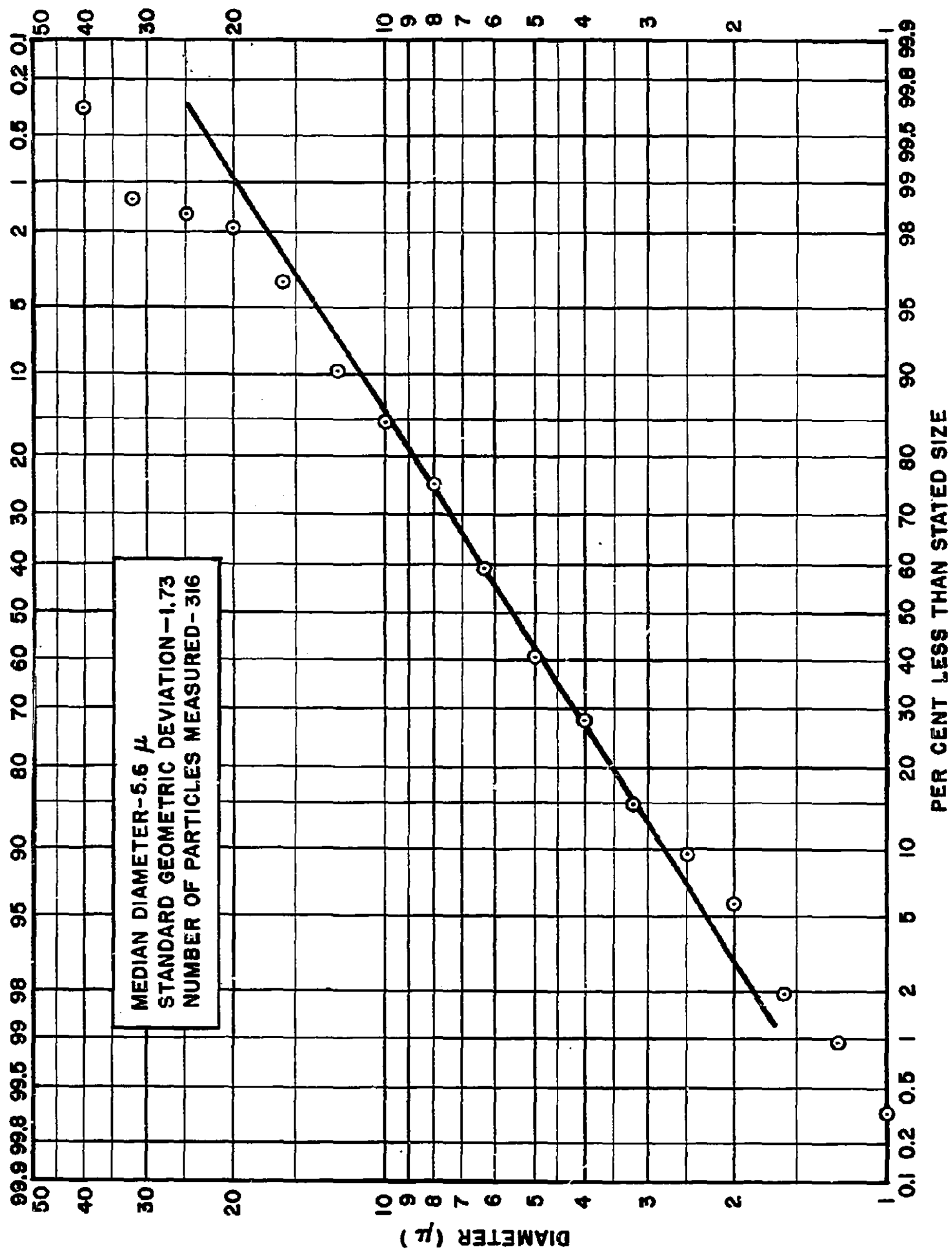


FIG. 3.3 Size Distribution of Individual Radioactive Spheres Found in Samples from Bogallua Island (Easy Shot)

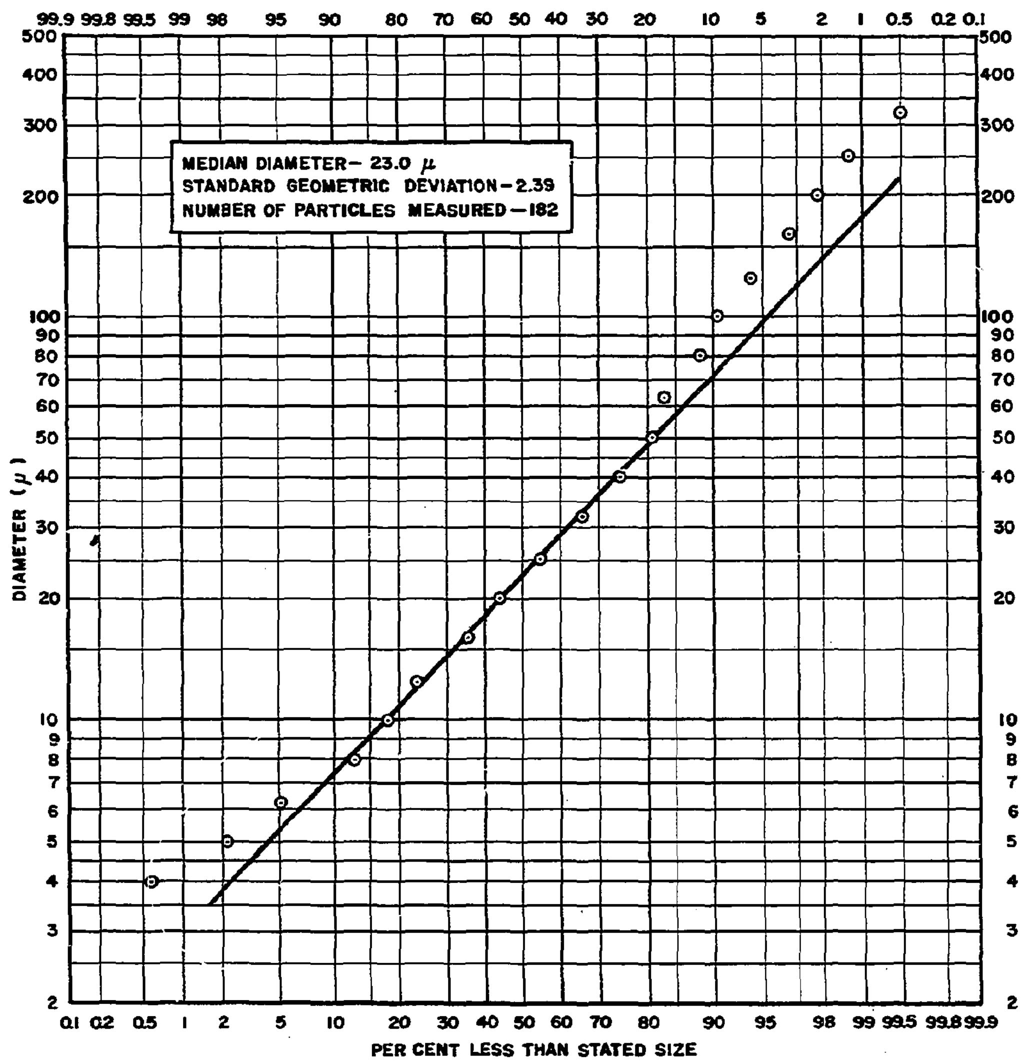


FIG. 3.4 Size Distribution of Radioactive Coral Grains Found in Samples from Rigili Island (Dog Shot)

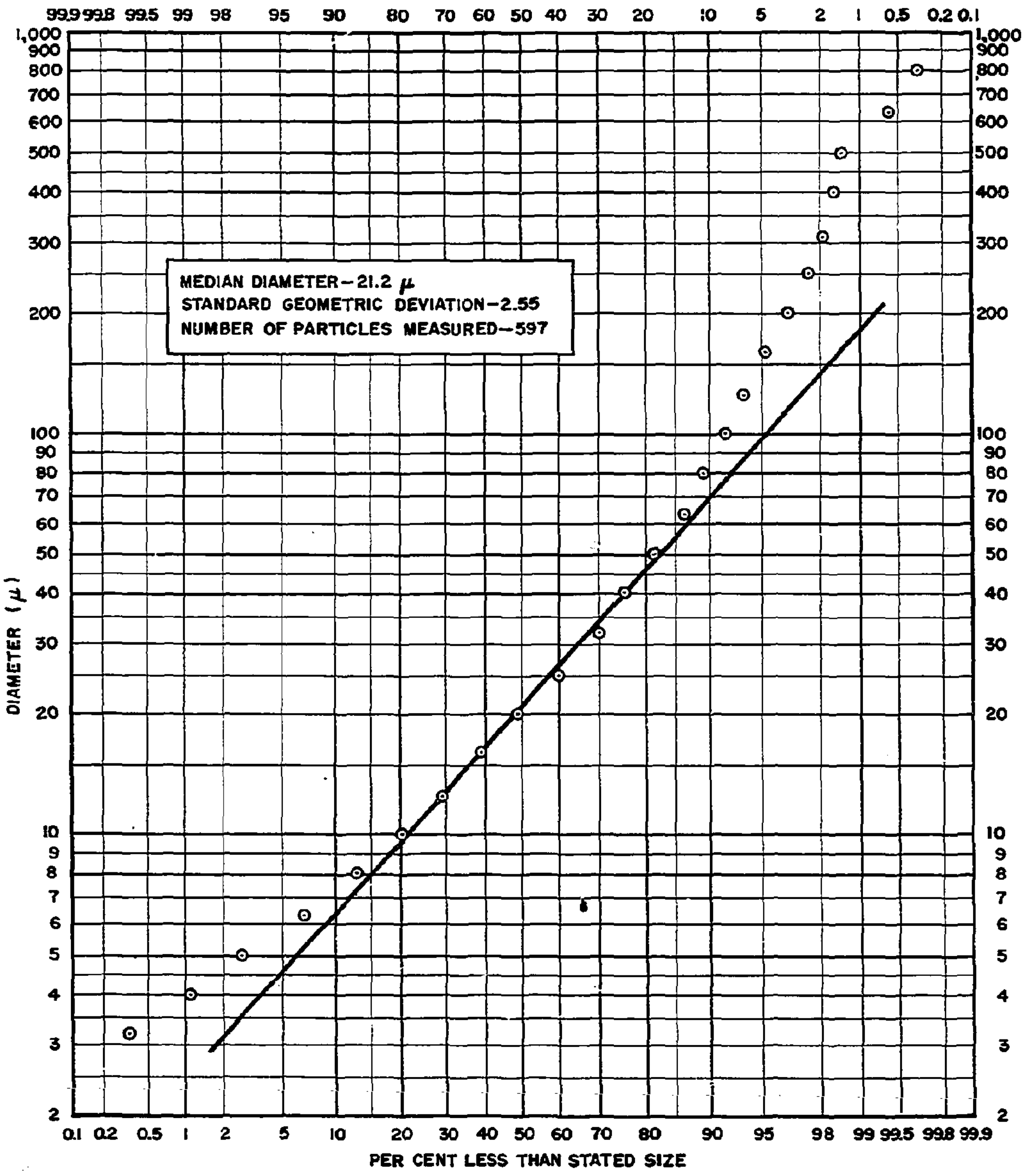


FIG. 3.5 Size Distribution of Radioactive Coral Grains Found in Samples from Bogallua Island (Easy Shot)

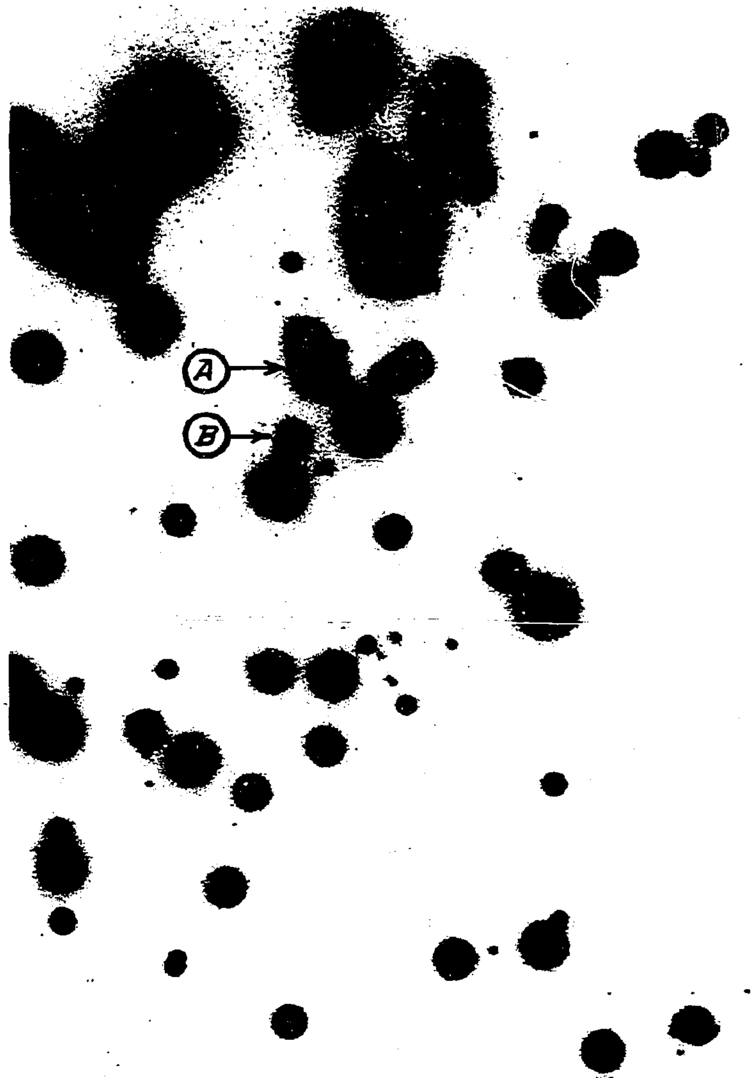


FIG. 3.6 Radioautograph of a Fall-out Plate from Parry Island (Dog Shot)